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(54) 【発明の名称】 積層品

(57)【要約】

【課題】本発明は、軽量、かつリサイクル性に優れ、成 形加工が容易で外観美麗な積層品を提供せんとするもの である。

【解決手段】本発明の積層品は、スキン剥離強度が20 N/c m以下であり、L値が60以下であるポリオレフィン系樹脂発泡体と、厚さが5 mm以上、密度が50 k g/c m^3 以下の嵩高性不織布とを一体成形してなる積層体であって、該積層体の目付が3 k g/ m^2 以下であることを特徴とするものである。

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【特許請求の範囲】

【請求項1】 スキン剥離強度が20N/cm以下であり、L値が60以下であることを特徴とするポリオレフィン系樹脂発泡体と、厚さが5mm以上、密度が50k g/ cm^3 以下の嵩高性不織布とを一体成形してなる積層体であって、該積層体の目付が3k g/ m^2 以下であることを特徴とする積層品。

【請求項2】 前記ポリオレフィン系樹脂発泡体において、ポリオレフィン系樹脂が、ポリエチレン系樹脂およびポリプロピレン系樹脂の少なくとも一つから選ばれる請求項1記載の積層品。

【発明の詳細な説明】

[0001]

【発明の属する技術分野】本発明は自動車の内装材、例 えば天井材、フロアーインシュレータ、ダッシュインシュレータ、リヤーパーテンションパネルインシュレーター、トランクトリム、ドアトリム等に関する。

[0002]

【従来の技術】近年、自動車の燃費向上のため、自動車 用内装材の軽量化が検討されており、また環境保護の観 20 点からリサイクル可能であることが切に要望されるよう になってきた。従来の自動車用内装材は塩化ビニルのシ ートとポリオレフィン系樹脂発泡体とを貼り合わせたも のを用いていたが、リサイクル等の観点から熱可塑性エ ラストマーのシートとポリオレフィン系樹脂発泡体とを 貼り合わせたものが用いられるようになってきた。ま た、自動車用内装材として、ポリオレフィン系樹脂発泡 体と不織布を貼り合わせたものも用いられている。しか し、ポリオレフィン系樹脂発泡体は剛性が低いため、例 えば自動車用天井材として用いた場合は垂れ下がるとい 30 う問題点が発生し、そのためガラス繊維や無機フィラー を添加することで補強したり、ガラス繊維からなるシー トを補強剤として積層する必要があり、このためリサイ クルが非常に困難であるという問題点があった。

[0003]

【発明が解決しようとする課題】本発明は、かかる従来 技術の背景に鑑み、軽量、かつリサイクル性に優れ、成 形加工が容易で外観美麗な積層品を提供せんとするもの である。

[0004]

【課題を解決するための手段】本発明は、かかる課題を解決するために、次のような手段を採用するものである。すなわち、本発明の積層品は、スキン剥離強度が 20N/cm以下であり、L値が60以下であるポリオレフィン系樹脂発泡体と、厚さが 5mm以上、密度が $50kg/cm^3$ 以下の嵩高性不織布とを一体成形してなる積層体であって、該積層体の目付が $3kg/m^2$ 以下であることを特徴とするものである。

[0005]

【発明の実施の形態】本発明は、前記課題、つまり軽

量、かつリサイクル性に優れ、成形加工が容易で外観美麗な積層品について、鋭意検討し、特定なポリオレフィン系樹脂発泡体と嵩高性不織布とを一体成形してなる特定な目付の積層体としてみたところ、かかる課題を一挙に解決することを究明したものである。

【0006】すなわち、本発明に用いるポリオレフィン系樹脂発泡体は、ポリオレフィン系樹脂100重量部に対し、有機系熱分解型発泡剤1~50重量部を所定形状に成形した後、架橋、発泡したものが使用される。

【0007】かかるポリオレフィン系樹脂としては、ポリエチレン系樹脂およびポリプロピレン系樹脂の少なくとも1種類を含有することが好ましく、これらポリオレフィン系樹脂を80重量%以上含有しているのが、リサイクル性、成形加工性ならびに外観美麗性の上から好ましい。

【0008】本発明で使用するポリプロピレン系樹脂としては、好ましくは分子量分布が $1\sim10$ 、MFRが 0. $5\sim15$ g /10 分であり、プロピレンの単独重合体或いはプロピレンとプロピレン以外の α - オレフィンとの共重合体であるものが好ましく使用される。かかる共重合成分の α - オレフィン含有量は、好ましくは $1\sim15$ 重量%であり、 α - オレフィンとしては、例えばエチレン、1 - ブテン、1 - ペンテン、1 - ペンテン、4 - メチル- 1 - ペンテン、1 - ペンテン、1 - ペンテン、1 - オクテン、1 - ペンテン、1 - ペンテン、1 - オクテン、1 - ペンテン、1 - オクテン、1 - ペンテン、1 - オクテン、1 - ペンテン、1 - ペンテン、1 - オクテン、1 - ペンテン、1 - ペンテン、1 - オクテン、1 - ペンテン、1 - オクテン、1 - ペンテン、1 - ペンテン、1 - オクテン、1 - ペンテン、1 - - ペンテン、1 - - - + センなどが好ましく使用される。

【0009】本発明で使用するポリエチレン系樹脂としては、好ましくは密度が $860\sim975$ k g/m³、MFRが0.5 ~15 g/10分であり、エチレンの単独重合体或いはエチレンと α -オレフィンとの共重合体であるものが好ましく使用される。かかる共重合成分の α -オレフィンとしては、例えば、プロピレン、1-ブテン、1-ペンテン、1-ペナルー1-ペンテン、1-ペプテン、1-ペナテン等が好ましく使用される。

【0010】本発明で使用する有機系熱分解型発泡剤としては、加熱により分解して気体を発生する化合物が好ましく使用され、具体的には、アゾジカルボンアミド、オキシベンゼンスルホニルヒドラジド、ジニトロソペンタメチレンテトラミン、トルエンスルホニルヒドラジド、4,4ーオキシビス(ベンゼンスルホニルヒドラジド)、アゾビスイソブチロニトリル、アゾジカルボン酸バリウム、ヒドラゾジカルボンアミド等が好ましく使用される。これらは単独で用いてもよいし、併用してもよい。有機系熱分解型発泡剤は、樹脂成分100重量部に対して、好ましくは1~50重量部の割合で使用する。有機系熱分解型発泡剤の添加量は、少なすぎると樹脂組成物の発泡性、すなわち軽量性が低下し、多すぎると得られる発泡体の強度が低下する。有機系へ熱分解型発泡50剤の好ましい使用割合は、4~30重量部である。

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【0011】本発明においては、本発明の目的を損なわ ない範囲内で、他の熱可塑性樹脂、例えば低密度ポリエ チレン、直鎖状低密度ポリエチレン、中密度ポリエチレ ン、高密度ポリエチレン、ポリプロピレン、エチレンー ポリプロピレンゴム、ポリ酢酸ビニル、ポリブテン等の 樹脂を少量成分として添加することができる。また、フ ェノール系、リン系、アミン系、イオウ系等の酸化防止 剤、多官能性モノマーであるジビニルベンゼン、トリメ チロールプロパントリメタクリレート、1,9-ノナン ジオールジメタクリレート、1,10-デカンジオール ジメタクリレート、トリアリルイソシアヌレート、エチ ルビニルベンゼン、エチレンビニルジメタクリレート、 1. 2-ベンゼンジカルボン酸ジアリルエステル、1. 3-ベンゼンジカルボン酸ジアリルエステル、1,4-ベンゼンジカルボン酸ジアリルエステル、1.2.4-ベンゼントリカルボン酸ジアリルエステル等の架橋助 剤、金属害防止剤、難燃剤、充填剤、帯電防止剤、安定 剤、顔料などを添加してもよい。

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【0012】本発明では、前記各成分を配合して得られたポリオレフィン系樹脂組成物を所定形状に成形した後、架橋・発泡して発泡体を製造する。具体的には、例えば、下記の製造方法が好ましく採用される。

【0013】前記ポリオレフィン系樹脂組成物の所定量を、単軸押出機、二軸押出機、バンバリーミキサー、ニーダーミキサー、ミキシングロール等の混練装置を用いて、熱分解型発泡剤の分解温度未満で均一に溶融混練し、これをシート状に成形する。次いで、得られたシートに電離性放射線を所定線量照射してオレフィン系樹脂を架橋させ、この架橋シートを熱分解型発泡剤の分解温度以上に加熱して発泡させる。電離性放射線照射による架橋にかえて、過酸化物による架橋や、シラン架橋を行っても良い。

【0014】かかる電離性放射線としては、 α 線、 β 線、y線、電子線等を挙げることができる。電離性放射線の照射線量は、多官能性モノマーの種類、添加量、目的とする架橋度等によって異なるが、好ましくは $1\sim5$ 00kGy、より好ましくは $5\sim300$ kGyである。この照射エネルギーによって生じせしめる架橋、すなわちゲル分率は、好ましくは $10\sim70$ %で、このゲル分率が10%未満では発泡時表面から発泡剤のガスが逸散し、表面があれたり、所望の発泡倍率の製品が得られにくくなるので好ましくなく、一方、70%を越えると過度の架橋となり高発泡品が得られなくなることと硬くなるため低温下での衝撃性が悪化し割れが生じやすくなるので好ましくない。

【0015】本発明の発泡体は各種の発泡法、具体的には縦(横)型熱風発泡法、薬液浴上発泡法などに例示される方法にて製造される。

【0016】本発明のポリオレフィン系樹脂発泡体における架橋度は、好ましくは10~70%、さらに好まし

くは15~50%であることが、架橋後シートの発泡安定性と発泡セルが細かく、均一に近い製品外観及び厚み均一性に優れたポリオレフィン系樹脂発泡体を得る上において好ましい。

【0017】なお、かかる架橋度は、発泡体を細断し約0.2g(W₀g)を精秤したものを溶媒としてキシレンを用い、120℃下のソックスレイ抽出器で24時間抽出後、不溶分を取り出し、純粋なキシレンで洗浄後、さらにアセトンで洗浄し80℃に加熱した真空乾燥機で4時間加熱、揮発分を完全に除去後、室温で自然冷却する。このものの重量(W₁g)を測定し、次式にて架橋度を求める。

【0018】架橋度=(W₁/W₀) x100(%) さらに、本発明のポリオレフィン系樹脂発泡体におけるスキン剥離強度は20N/cm以下であることが好ましく、より好ましくは15N/cm以下である。スキン剥離強度が20N/cm以上あると、貼り合わせた不織布を剥離する際に、材質強度の弱い部分が凝集破壊してしまい、不織布に凝集破壊したポリオレフィン系樹脂発泡体が付着したり、逆にポリオレフィン系樹脂発泡体が付着したり、逆にポリオレフィン系樹脂発泡体に凝集破壊した不織布が付着してしまいリサイクルが困難となるので好ましくない。

【0019】本発明で使用するポリオレフィン系樹脂発 泡体のL値は、好ましくは60以下、より好ましくは5 0以下である。L値が60より大きいと、車体に取り付 けた後、汚れなどが目立ちやすく、外観上好ましくな

【0020】かかるL値とは、色相を表す尺度であり、本発明においてはカラーマシン(スガ試験機株式会社製SMカラーコンピューター SM-5型)にて測定したものである。

【0021】本発明の積層品は、嵩高性不織布とポリオレフィン系樹脂発泡体とを一体成形してなるが、その貼り合わせには接合剤を用いても、用いなくてもよい。リサイクル性からは、接合剤を用いずに貼り合わせるのが好ましい。積層品の貼り合わせの際に用いられる接合剤としては、加熱溶融し反応固化するフェノール樹脂やアミノ樹脂、ウレタン系、合成ゴム系、エポキシ系の接着剤、ポリエチレンやポリプロピレンの樹脂或いは繊維等が挙げられ、これらは単独で用いても2種類以上を併用して用いても良い。

【0022】一方、接合剤を用いない場合に不織布と発泡体を貼り合わせることが出来るのは、現段階では必ずしも明確ではないが、以下のように推定される。嵩高性不織布とポリオレフィン系架橋樹脂発泡体とを一体成形する際にはどちらか一方、好ましくは両方のシートを加熱する。これらを貼り合わせ、プレスにより一体成形すると、嵩高性不織布の表層部分がポリオレフィン系樹脂発泡体の表層部分にめり込み、すなわちアンカー効果的なものが発現し、両者が接合剤を用いることなく貼り合

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わせられるものと考えられる。

【0023】本発明で用いる嵩高性不織布の厚みは5m m以上、密度が50kg/cm²以下であることが好ま しい。厚みが5mmより薄いと成形品の剛性が不足し、 例えば天井材などに用いた場合は垂れ等が生じるため好 ましくなく、密度が50kg/cm3以上であるとポリ オレフィン系樹脂発泡体との貼り合わせにおいて不織布 がポリオレフィン系樹脂発泡体の表層部分にめり込みに くくなり、貼り合わせの強度が弱くなる。

【0024】本発明で使用するポリオレフィン系樹脂発 泡体のスキン剥離強度は、以下のような方法で測定を行 った。ポリオレフィン系樹脂発泡体と厚み1mm、目付 1. 36 k g/m² の塩化ビニルのシートを接着剤で貼 り合わせ、幅25mm及び長さ100mmの寸法の試験 片を作成する。これをオートグラフ(株式会社オリエン テック製「UCT500」)を用いて、室温で塩化ビニ ルシートを剥離させたときの強度を測定したものであ る。

[0025]

【実施例】次に本発明を実施例に基づいて説明するが、 本発明はこれに限定されるものではない。以下「部」と あるのは「重量部」を意味する。

【0026】実施例1

(1) ポリオレフィン系樹脂発泡体の製造

ポリエチレン系樹脂として、MFRが5g/10分であ り、融点が125℃のポリエチレン50部、ポリプロピ レン系樹脂として、MFRが2.5g/10分、エチレ ンが4.2%ランダム共重合したエチレンープロピレン 共重合体50部、架橋助剤としてジビニルベンゼン3 部、熱分解型発泡剤としてアゾジカルボンアミド12 部、酸化防止剤として2, 6-ジーt-ブチルーp-ク レゾール0.3部、顔料としてカーボンブラック0.1 部を2軸押し出し機に供給し、温度180℃で溶融混練 し、厚さ1.5mmの発泡性ポリオレフィン系樹脂シー トを得た。得られた発泡性ポリオレフィン系樹脂シート に、加速電圧800kVで電離性放射線として電子線を 300kGy照射し架橋させ、発泡性ポリオレフィン系 樹脂架橋シートを得た。得られた発泡性ポリオレフィン 系樹脂架橋シートを縦型発泡炉に供給し、炉内温度24 5℃で連続的に延伸し、加熱発泡させてポリオレフィン 40 系樹脂発泡体を得た。このようにして得られた発泡体は 厚み3.0mm、架橋度35%、発泡倍率25倍、L 値が41、スキン剥離強度が8.2N/cmのものであ った。

(2) 積層体の製造

得られたポリオレフィン系樹脂発泡体をラジエーション ヒータで表面温度が145℃になるまで加熱し、表面を 170℃になるまで加熱した厚さ50mm、密度25k g/m³であるポリエステルの不織布と貼り合わせ一体成 形し、積層体を得た。

【0027】比較例1

実施例で示したポリオレフィン系樹脂発泡体の製造にお いて、カーボンブラックを添加しないこと以外は実施例 1と同様の操作を行い、厚み3.0mm、架橋度35 %、発泡倍率25倍、L値が75のポリオレフィン系樹 脂発泡体を得、さらに実施例と同様の操作を行い、積層 体を得た。

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【0028】比較例2

実施例で示したポリエステルの不織布の代わりに、厚さ 50mm、密度75kg/m³のポリエステル不織布を用 いた以外は実施例と同様の操作を行い、積層体を得た。

【0029】比較例3

実施例で示したポリオレフィン系樹脂発泡体のかわり に、厚3.0mm、発泡倍率が5倍、架橋度35%、L 値38、スキン剥離強度23.2N/cmのポリオレフ ィン系樹脂発泡体を用いた以外は実施例と同様の操作を 行い、積層体を得た。

【0030】上記実施例及び比較例1,2、3において 得られた積層体について、剥離強度を測定し嵩高性不織 20 布との接着性を評価し、得られた結果を表1に示した。 外観性の判定は、積層品のポリオレフィン系樹脂発泡体 に黒インクを滴下し、それが目立つ場合を×、そうでな い場合を○とした。

【0031】剥離状態の判定は、室温で発泡体を剥離さ せ、剥離したポリオレフィン系樹脂発泡体に嵩高性不織 布の繊維の付着、もしくは嵩高性不織布の表面に凝集破 壊したポリオレフィン系樹脂発泡体が確認された場合を ×、付着が確認されなかった場合を○とした。

[0032]

【表1】

30

| İ | 外観性 | 剥離強度 | 剥離状態 | |
|-------|-----|--------|------|--|
| | | (N/cm) | | |
| 実施例 1 | 0 | 0.304 | 0 | |
| 比較例1 | × | 0.304 | 0 | |
| 比較例 2 | 0 | 測定不能* | | |
| 比較例 3 | 0 | 2.99 | × | |

【0033】表中、*は測定限界以下であることを示

【0034】表1から明らかなように、実施例のもの は、比較例に比して、軽量で、成形加工が容易で、外観 の美麗な積層品であり、リサイクル性にも優れるもので あった。

[0035]

【発明の効果】本発明によれば、軽量で、リサイクル性 に優れ、成形加工が容易な上に、外観美麗な積層品を再 現性よく提供することができる。

フロントページの続き

Fターム(参考) 3D023 BA01 BA02 BB02 BB08 BB17 BD01 BD03 BD12 BE04 4F100 AA37A AK03A AK04A AK07A AK41B AK64A AL05A BA02 CA01A CA02A CA13A DG15B DJ01A EJ02 EJ37 EJ53 GB07 GB33 JA13 JA13B JK06A JL03 JL16 YY00A YY00B

[Claim(s)]

[Claim 1] For thickness, 5mm or more and a consistency are [the polyolefine system resin foam characterized by for skin peel strength being 20 or less N/cm, and L value being 60 or less, and] 50 kg/cm3. It is the layered product which really comes to fabricate the following loft nonwoven fabrics, and the eyes of this layered product are 3 kg/m2. Laminate characterized by being the following.

[Claim 2] The laminate according to claim 1 with which polyolefine system resin is chosen from at least one of polyethylene system resin and the polypropylene regins in said polyolefine system resin foam.

DETAILED DESCRIPTION

[Detailed Description of the Invention] [0001]

[Field of the Invention] This invention relates to the interior material of an automobile, for example, head-lining material, a floor insulator, a dash insulator, a RIYAPA tension panel insulator, a trunk trim, a door trim, etc.

[0002]

[Description of the Prior Art] In recent years, to consider lightweight-ization of the interior material for automobiles, and to be able to recycle from a viewpoint of environmental protection has come to be eagerly demanded for the improvement in fuel consumption of an automobile. Although the conventional interior material for automobiles used what stuck the sheet of a vinyl chloride, and polyolefine system resin foam, what stuck the sheet of thermoplastic elastomer and polyolefine system resin foam from viewpoints, such as recycle, has come to be used. Moreover, what stuck polyolefine system resin foam and a nonwoven fabric is used as interior material for automobiles. However, when it used as head-lining material for automobiles since polyolefine system resin foam has low rigidity for example, it needed to reinforce with the trouble of hanging down occurring, therefore adding a glass fiber and an inorganic filler, or it needed to carry out the laminating, having used as the reinforcing agent the sheet which consists of a glass fiber, and, for this reason, there was a trouble that recycle was very difficult.

[0003]

[Problem(s) to be Solved by the Invention] this invention -- the background of this conventional technique -- taking an example -- a light weight and recycle nature -- excelling -- fabrication -- easy -- an appearance -- a beautiful laminate -- it is going to provide -- it is a thing.

[0004]

[Means for Solving the Problem] The following means are used for this invention in order to solve this technical problem. That is, for thickness, 5mm or more and a consistency are [the laminate of this invention / the polyolefine system resin foam whose skin peel strength is 20 or less N/cm and whose L value is 60 or less, and] 50 kg/cm3. It is the layered product which really comes to fabricate the following loft nonwoven fabrics, and the eyes of this layered product are 3 kg/m2. It is characterized

by being the following. [0005]

[Embodiment of the Invention] this invention -- said technical problem, i.e., a light weight, and recycle nature -- excelling -- fabrication -- easy -- an appearance -- a beautiful laminate is examined wholeheartedly, and when it sees as a layered product of the eyes [****] which really come to fabricate polyolefine system resin foam [****] and a loft nonwoven fabric, it studies solving this technical problem at once.

[0006] That is, that to which the polyolefine system resin foam used for this invention constructed the bridge over after fabricating the organic system pyrolysis mold foaming agent 1-50 weight sections in a predetermined configuration to the polyolefine system resin 100 weight section, and it foamed is used.

[0007] As this polyolefine system resin, it is desirable to contain at least one kind of polyethylene system resin and a polypropylene regin, and it is desirable from recycle nature, fabrication nature, and appearance beautiful nature to contain these polyolefine system resin 80% of the weight or more.

[0008] As a polypropylene regin used by this invention, the thing 1-10, and whose MFR are 0.5-15g / 10 minutes and whose molecular weight distribution are the homopolymer of a propylene or a copolymer with alpha olefins other than a propylene and a propylene is used preferably. The alpha olefin content of this copolymerization component is 1 - 15 % of the weight preferably, and an ethylene, 1-butene, 1-pentene, 1-hexene, 4-methyl-1-pentene, 1-heptene, 1-octene, 3, and 3-dimethyl-1-hexene etc. is preferably used as an alpha olefin, for example.

[0009] As polyethylene system resin used by this invention, the thing 860 - 975 kg/m3 and whose MFR are 0.5 - 15 g / 10 minutes and whose consistency is the homopolymer of ethylene or the copolymer of ethylene and an alpha olefin is used preferably. As an alpha olefin of this copolymerization component, a propylene, 1-butene, 1-pentene, 1-hexene, 4-methyl-1-pentene, 1-heptene, 1-octene, etc. are used preferably, for example.

[0010] The compound which decomposes with heating and generates a gas as an organic system pyrolysis mold foaming agent used by this invention is used preferably, specifically, an AZOJI carvone amide, oxybenzene sulfonylhydrazide, dinitrosopentamethylenetetramine. tosyl hydrazide, 4, and 4-oxy-screw (benzenesulphonyl hydrazide), azobisisobutyronitril, azo dicarboxylic acid barium, a hydrazo JIKARUBON amide, etc. are used preferably. These may be used independently and may be used together. An organic system pyrolysis mold foaming agent is preferably used at a rate of 1 - 50 weight section to the resinous principle 100 weight section. If there are too few additions of an organic system pyrolysis mold foaming agent, the fizz of a resin constituent, i.e., lightweight nature, will fall, and the reinforcement of the foam which will be obtained if many [too] falls. The desirable operating rate of a pyrolysis mold foaming agent is 4 - 30 weight section in an organic system.

[0011] In this invention, resin, such as other thermoplastics, for example, low density polyethylene, straight chain-like low density polyethylene, medium density polyethylene, high density polyethylene, polypropylene, ethylene-polypropylene rubber, polyvinyl acetate, and polybutene, can be added as a little component within

limits which do not spoil the purpose of this invention. Moreover, antioxidants, such as a phenol system, the Lynn system, an amine system, and a sulfur system, The divinylbenzene, trimethylolpropanetrimethacrylate which are a polyfunctional monomer, 1, 9-nonane diol dimethacrylate, 1, 10-Deccan diol dimethacrylate, Triallyl isocyanurate, ethyl vinylbenzene, ethylene vinyl dimethacrylate, 1, 2-benzene dicarboxylic acid diallyl ester, Bridge formation assistants, such as 1, 4-benzene dicarboxylic acid diallyl ester, and 1,2,4-benzenetricarboxylic acid diallyl ester, a metal damage inhibitor, a flame retarder, a bulking agent, an antistatic agent, a stabilizer, a pigment, etc. may be added.

[0012] In this invention, after fabricating the polyolefine system resin constituent which blended said each component and was obtained in a predetermined configuration, it constructs a bridge and foams and foam is manufactured. Specifically, the following manufacture approach is adopted preferably.

[0013] Melting kneading of the specified quantity of said polyolefine system resin constituent is carried out under with the decomposition temperature of a pyrolysis mold foaming agent at homogeneity using kneading equipments, such as a single screw extruder, a twin screw extruder, a Banbury mixer, a kneader mixer, and a roll mill, and this is fabricated in the shape of a sheetSubsequently, carry out the predetermined dosage exposure of the ionizing radiation, and olefin system resin is made to construct a bridge over the obtained sheet, and this bridge formation sheet is heated more than the decomposition temperature of a pyrolysis mold foaming agent, and is made to foam. It may change to bridge formation by ionizing radiation exposure, and bridge formation by the peroxide and silane bridge formation may be performed.

[0014] As this ionizing radiation, alpha rays, beta rays, a gamma ray, an electron ray, etc. can be mentioned. Although the quantity of radiation of an ionizing radiation changes with the class of polyfunctional monomer, an addition, degrees of cross linking made into the purpose, it is 5-300kGy more preferably one to 500 kGy. The bridge formation out of which it produces and cheats with this exposure energy, i.e., a gel molar fraction Are 10 - 70% preferably and the gas of a foaming agent carries out [this gel molar fraction] fly off from a front face at less than 10% at the time of foaming. Since the product of desired expansion ratio becomes there needs to be a front face enough and is hard to be obtained, it will become too much bridge formation and it will become hard with a high foaming article no longer being obtained, if 70% is exceeded on the other hand preferably, and the impact nature under low temperature gets worse and it becomes easy to produce a crack, it is not desirable.

[0015] The foam of this invention is manufactured by various kinds of foaming methods and the approach specifically illustrated by the vertical (width) mold hot blast foaming method, the drug solution bath top foaming method, etc.

[0016] 10 to 70%, the foaming stability and the foaming cel of the sheet after bridge formation are fine, that it is 15 - 50% still more preferably obtains polyolefine system resin foam excellent in the product appearance near homogeneity, and thickness homogeneity upwards, and the degree of cross linking in the polyolefine system resin foam of this invention has it. [preferably desirable] [0017] In addition, 4-hour heating and volatile matter are completely cooled naturally after removal and at a room temperature with the vacuum dryer which took out after a 24-hour extract and insoluble matter with the Soxhlet extractor under 120 degrees C using the xylene by having

used as the solvent what this degree of cross linking carried out beating of the foam, and weighed about 0.2g (W0g) precisely, washed with the acetone further after washing by the pure xylene, and was heated at 80 degrees C. The weight (W1g) of this thing is measured and it asks for a degree of cross linking in a degree type.

[0018] Degree of cross linking = $(W1/W0) \times 100 (\%)$

Furthermore, as for the skin peel strength in the polyolefine system resin foam of this invention, it is desirable that they are 20 or less N/cm, and they are 15 or less N/cm more preferably. If there is 20 or more N/cm of skin peel strength, in case the stuck nonwoven fabric will be exfoliated, since the polyolefine system resin foam which the part with weak quality-of-the-material reinforcement carried out [foam] cohesive failure, and carried out cohesive failure to the nonwoven fabric adheres, or the nonwoven fabric which carried out cohesive failure to polyolefine system resin foam conversely adheres and recycle becomes difficult, it is not desirable.

[0019] L value of the polyolefine system resin foam used by this invention is 50 or less more preferably 60 or less. dirt if L value is larger than 60, after attaching in a car body etc. -- conspicuous -- easy -- an exterior -- it is not desirable.

[0020] This L value is a scale showing a hue, and it measures in this invention by the color machine (SM color computer [by Suga Test Instruments Co., Ltd.] SM-5 mold).

[0021] Although it really comes to fabricate a loft nonwoven fabric and polyolefine system resin foam, even if it uses cement for the lamination, it is not necessary to use the laminate of this invention. It is desirable to stick without using cement from recycle nature. As cement used in the case of the lamination of a laminate, heating fusion is carried out, and the adhesives of phenol resin, amino resin, an urethane system, a synthetic-rubber system, and an epoxy system, polyethylene, resin or fiber of polypropylene etc. that carries out reaction solidification may be mentioned, and these may be used independently, or may use together and use two or more kinds. [0022] At a present stage, it is presumed as follows that a nonwoven fabric and foam can be stuck on the other hand when not using cement, although it is not necessarily clear. In case a loft nonwoven fabric and polyolefine system bridge formation resin foam are really fabricated, on the other hand, both sheets are preferably heated in which. If these are really fabricated with lamination and a press, the surface part of a loft nonwoven fabric will sink into the surface part of polyolefine system resin foam, namely, an anchor effect-thing will be discovered, and it will be thought that it is stuck without both using cement.

[0023] For the thickness of the loft nonwoven fabric used by this invention, 5mm or more and a consistency are 50 kg/cm3. It is desirable that it is the following. Since a lappet etc. arises when thickness was thinner than 5mm, and the rigidity of mold goods runs short, for example, it uses for head-lining material etc., consistencies are 50 kg/cm3 preferably. In lamination with polyolefine system resin foam, a nonwoven fabric stops easily being able to sink into the surface part of polyolefine system resin foam as it is above, and the reinforcement of lamination becomes weak.

[0024] The skin peel strength of the polyolefine system resin foam used by this invention measured by the following approaches. Polyolefine system resin foam, the thickness of 1mm, and eyes 1.36~kg/m2 The test piece of a dimension with lamination, a width of face [of 25mm], and a die length of 100mm is created for the sheet of a vinyl chloride with adhesives. The reinforcement at the time of making a chlorination vinyl sheet this exfoliate at a room temperature using an autograph (product made from an incorporated company cage en tech "UCT500") is measured.

[Example] Next, although this invention is explained based on an example, this invention is not limited to this. That it is with the "section" below means the "weight section."

[0026] As manufacture polyethylene system resin of example 1(1) polyolefine system resin foam As the polyethylene 50 section whose MFRs are 5g / 10 minutes and whose melting point is 125 degrees C, and a polypropylene regin The ethylene propylene rubber 50 section in which ethylene carried out [MFR] random copolymerization 4.2% for 2.5g / 10 minutes, As a bridge formation assistant, as the divinylbenzene 3 section and a pyrolysis mold foaming agent The AZOJI carvone amide 12 section, The 2,6-di-t-butyl-p-cresol 0.3 section was supplied as an anti-oxidant, the carbon black 0.1 section was supplied to the biaxial extruder as a pigment, melting kneading was carried out at the temperature of 180 degrees C, and the fizz polyolefine system resin sheet with a thickness of 1.5mm was obtained. Carry out a 300kGy exposure, the electron ray was made to construct a bridge over the obtained fizz polyolefine system resin sheet as an ionizing radiation with the acceleration voltage of 800kV, and the fizz polyolefine system resin bridge formation sheet was obtained. The vertical mold foaming furnace was supplied, whenever [furnace temperature], the obtained fizz polyolefine system resin bridge formation sheet is continuously extended at 245 degrees C, and carried out heating foaming at them, and polyolefine system resin foam was obtained. Thus, one 25 times the expansion ratio [the thickness of 3.0mm, degree-of-cross-linking 35 %, and] of this and L value were [41 and the skin peel strength of the obtained foam] the things of 8.2 N/cm.

(2) it the nonwoven fabric of 50mm in thickness which heated the manufacture profit **** polyolefine system resin foam of a layered product until skin temperature became 145 degrees C at the radiation heater, and heated the front face until it became 170 degrees C, and the polyester which is consistency 25 kg/m3, and really [lamination] fabricated, and the layered product was obtained.

[0027] In the manufacture of polyolefine system resin foam shown in the example of comparison 1 example, except not adding carbon black, the same actuation as an example 1 was performed, and one 25 times the expansion ratio [the thickness of 3.0mm, 35% of degrees of cross linking, and] of this and L value obtained the polyolefine system resin foam of 75, performed the still more nearly same actuation as an example, and obtained the layered product.

[0028] Instead of the nonwoven fabric of the polyester shown in the example of comparison 2 example, except having used the polyester nonwoven fabric of 50mm in thickness, and consistency 75 kg/m3, the same actuation as an example was performed and the layered product was obtained. [0029] instead of [of the polyolefine system resin foam shown in the example of comparison 3 example] -- thick -- except that 3.0mm and expansion ratio used the polyolefine system resin foam of 5 times, 35% of degrees of cross linking, the L value 38, and skin peel strength 23.2 N/cm, the same actuation as an example was performed and the layered product was obtained.

[0030] About the layered product obtained in the above-mentioned example and the examples 1, 2, and 3 of a comparison, peel strength was measured, the adhesive property with a loft nonwoven fabric was evaluated, and the obtained result was shown in Table 1. The judgment of appearance nature trickled black ink into the polyolefine system resin foam of a laminate, made the case where it was conspicuous as x, and made 0 the case where that was not right.

[0031] The judgment of a desquamative state made O the case where x and adhesion were not checked in the case where the polyolefine system resin foam which carried out cohesive failure to the polyolefine system resin foam which foam was made to exfoliate and exfoliated at the room temperature on adhesion of the fiber of a loft nonwoven fabric or the front face of a loft nonwoven

. fabric is checked. [0032]
[Table 1]

| | 外観性 | 剥離強度 | 剥離状態 |
|-------|-----|--------|------|
| | | (N/cm) | |
| 実施例 1 | 0 | 0.304 | 0 |
| 比較例1 | × | 0.304 | 0 |
| 比較例 2 | 0 | 測定不能* | - |
| 比較例 3 | 0 | 2.99 | × |

[0033] It is shown that front Naka and * are below measurement limitations.

[0034] The thing of an example was what it is lightweight, and fabrication is easy, is the beautiful laminate of an appearance, and is excellent also in recycle nature as compared with the example of a comparison so that clearly from Table 1.

[0035]

[Effect of the Invention] according to this invention -- lightweight -- recycle nature -- excelling -- the top where fabrication is easy -- an appearance -- a beautiful laminate can be offered with sufficient repeatability.

TECHNICAL FIELD

[Field of the Invention] This invention relates to the interior material of an automobile, for example, head-lining material, a floor insulator, a dash insulator, a RIYAPA tension panel insulator, a trunk trim, a door trim, etc.

PRIOR ART

[Description of the Prior Art] In recent years, to consider lightweight-ization of the interior material for automobiles, and to be able to recycle from a viewpoint of environmental protection has come to be eagerly demanded for the improvement in fuel consumption of an automobile. Although the conventional interior material for automobiles used what stuck the sheet of a vinyl chloride, and polyolefine system resin foam, what stuck the sheet of thermoplastic elastomer and polyolefine system resin foam from viewpoints, such as recycle, has come to be used. Moreover, what stuck polyolefine system resin foam and a nonwoven fabric is used as interior material for automobiles. However, when it used as head-lining material for automobiles since polyolefine system resin foam has low rigidity for example, it needed to reinforce with the trouble of hanging down occurring, therefore adding a glass fiber and an inorganic filler, or it needed to carry out the laminating, having used as the reinforcing agent the sheet which consists of a glass fiber, and, for this reason, there was a trouble that recycle was very difficult.

EFFECT OF THE INVENTION

[Effect of the Invention] according to this invention -- lightweight -- recycle nature -- excelling -- the top where fabrication is easy -- an appearance -- a beautiful laminate

[Problem(s) to be Solved by the Invention] this invention -- the background of this conventional technique -- taking an example -- a light weight and recycle nature -- excelling -- fabrication -- easy -- an appearance -- a beautiful laminate -- it is going to provide -- it is a thing.

MEANS

[Means for Solving the Problem] The following means are used for this invention in order to solve this technical problem. That is, for thickness, 5mm or more and a consistency are [the laminate of this invention / the polyolefine system resin foam whose skin peel strength is 20 or less N/cm and whose L value is 60 or less, and] 50 kg/cm3. It is the layered product which really comes to fabricate the following loft nonwoven fabrics, and the eyes of this layered product are 3 kg/m2. It is characterized by being the following.

[0005]

[Embodiment of the Invention] this invention -- said technical problem, i.e., a light weight, and recycle nature -- excelling -- fabrication -- easy -- an appearance -- a beautiful laminate is examined wholeheartedly, and when it sees as a layered product of the eyes [****] which really come to fabricate polyolefine system resin foam [****] and a loft nonwoven fabric, it studies solving this technical problem at once.

[0006] That is, that to which the polyolefine system resin foam used for this invention constructed the bridge over after fabricating the organic system pyrolysis mold foaming agent 1 - 50 weight sections in a predetermined configuration to the polyolefine system resin 100 weight section, and it foamed is used.

[0007] As this polyolefine system resin, it is desirable to contain at least one kind of polyethylene system resin and a polypropylene regin, and it is desirable from recycle nature, fabrication nature, and appearance beautiful nature to contain these polyolefine system resin 80% of the weight or more.

[0008] As a polypropylene regin used by this invention, the thing 1-10, and whose MFR are 0.5-15g / 10 minutes and whose molecular weight distribution are the homopolymer of a propylene or a copolymer with alpha olefins other than a propylene and a propylene is used preferably. The alpha olefin content of this copolymerization component is 1 - 15 % of the weight preferably, and an ethylene, 1-butene, 1-pentene, 1-hexene, 4-methyl-1-pentene, 1-heptene, 1-octene, 3, and 3-dimethyl-1-hexene etc. is preferably used as an alpha olefin, for example.

[0009] As polyethylene system resin used by this invention, the thing 860 - 975 kg/m3 and whose MFR are 0.5-15g / 10 minutes and whose consistency is the homopolymer of ethylene or the copolymer of ethylene and an alpha olefin is used preferably. As an alpha olefin of this copolymerization component, a propylene, 1-butene, 1-pentene, 1-hexene, 4-methyl-1-pentene, 1-heptene, 1-octene, etc. are used preferably, for example.

[0010] The compound which decomposes with heating and generates a gas as an organic system pyrolysis mold foaming agent used by this invention is used preferably,

specifically, an AZOJI carvone amide, oxybenzene sulfonylhydrazide, , and, hydrazide, 4, and 4-oxy-screw dinitrosopentamethylenetetramine, tosyl (benzenesulphonyl hydrazide), azobisisobutyronitril, azo dicarboxylic acid barium, a hydrazo JIKARUBON amide, etc. are used preferably. These may be used independently and may be used together. An organic system pyrolysis mold foaming agent is preferably used at a rate of 1 - 50 weight section to the resinous principle 100 weight section. If there are too few additions of an organic system pyrolysis mold foaming agent, the fizz of a resin constituent, i.e., lightweight nature, will fall, and the reinforcement of the foam which will be obtained if many [too] falls. The desirable operating rate of a pyrolysis mold foaming agent is 4 - 30 weight section in an organic system.

[0011] In this invention, resin, such as other thermoplastics, for example, low density polyethylene, straight chain-like low density polyethylene, medium density polyethylene, high density polyethylene, polypropylene, ethylene-polypropylene rubber, polyvinyl acetate, and polybutene, can be added as a little component within limits which do not spoil the purpose of this invention. Moreover, antioxidants, such as a phenol system, the Lynn system, an amine system, and a sulfur system, The divinylbenzene, trimethylolpropanetrimethacrylate which are a polyfunctional monomer, 1, 9-nonane diol dimethacrylate, 1, 10-Deccan diol dimethacrylate, Triallyl isocyanurate, ethyl vinylbenzene, ethylene vinyl dimethacrylate, 1, 2-benzene dicarboxylic acid diallyl ester, 1, 3-benzene dicarboxylic acid diallyl ester, Bridge formation assistants, such as 1, 4-benzene dicarboxylic acid diallyl ester, and 1,2,4-benzenetricarboxylic acid diallyl ester, a metal damage inhibitor, a flame retarder, a bulking agent, an antistatic agent, a stabilizer, a pigment, etc. may be added.

[0012] In this invention, after fabricating the polyolefine system resin constituent which blended said each component and was obtained in a predetermined configuration, it constructs a bridge and foams and foam is manufactured. Specifically, the following manufacture approach is adopted preferably.

[0013] Melting kneading of the specified quantity of said polyolefine system resin constituent is carried out under with the decomposition temperature of a pyrolysis mold foaming agent at homogeneity using kneading equipments, such as a single screw extruder, a twin screw extruder, a Banbury mixer, a kneader mixer, and a roll mill, and this is fabricated in the shape of a sheet. Subsequently, carry out the predetermined dosage exposure of the ionizing radiation, and olefin system resin is made to construct a bridge over the obtained sheet, and this bridge formation sheet is heated more than the decomposition temperature of a pyrolysis mold foaming agent, and is made to foam. It may change to bridge formation by ionizing radiation exposure, and bridge formation by the peroxide and silane bridge formation may be performed. [0014] As this ionizing radiation, alpha rays, beta rays, a gamma ray, an electron ray, etc. can be mentioned. Although the quantity of radiation of an ionizing radiation changes with the class of polyfunctional monomer, an addition, degrees of cross linking made into the purpose, it is 5-300kGy more preferably one to 500 kGy. The bridge formation out of which it produces and cheats with this exposure energy, i.e., a gel molar fraction Are 10 - 70% preferably and the gas of a foaming agent carries out

[this gel molar fraction] fly off from a front face at less than 10% at the time of foaming. Since the product of desired expansion ratio becomes there needs to be a front face enough and is hard to be obtained, it will become too much bridge formation and it will become hard with a high foaming article no longer being obtained, if 70% is exceeded on the other hand preferably, and the impact nature under low temperature gets worse and it becomes easy to produce a crack, it is not desirable.

[0015] The foam of this invention is manufactured by various kinds of foaming methods and the approach specifically illustrated by the vertical (width) mold hot blast foaming method, the drug solution bath top foaming method, etc.

[0016] 10 to 70%, the foaming stability and the foaming cel of the sheet after bridge formation are fine, that it is 15 - 50% still more preferably obtains polyolefine system resin foam excellent in the product appearance near homogeneity, and thickness homogeneity upwards, and the degree of cross linking in the polyolefine system resin foam of this invention has it. [preferably desirable]

[0017] In addition, 4-hour heating and volatile matter are completely cooled naturally after removal and at a room temperature with the vacuum dryer which took out after a 24-hour extract and insoluble matter with the Soxhlet extractor under 120 degrees C using the xylene by having used as the solvent what this degree of cross linking carried out beating of the foam, and weighed about 0.2g (W0g) precisely, washed with the acetone further after washing by the pure xylene, and was heated at 80 degrees C. The weight (W1g) of this thing is measured and it asks for a degree of cross linking in a degree type.

[0018] Degree of cross linking = $(W1/W0) \times 100 (\%)$

Furthermore, as for the skin peel strength in the polyolefine system resin foam of this invention, it is desirable that they are 20 or less N/cm, and they are 15 or less N/cm more preferably. If there is 20 or more N/cm of skin peel strength, in case the stuck nonwoven fabric will be exfoliated, since the polyolefine system resin foam which the part with weak quality-of-the-material reinforcement carried out [foam] cohesive failure, and carried out cohesive failure to the nonwoven fabric adheres, or the nonwoven fabric which carried out cohesive failure to polyolefine system resin foam conversely adheres and recycle becomes difficult, it is not desirable.

[0019] L value of the polyolefine system resin foam used by this invention is 50 or less more preferably 60 or less. dirt if L value is larger than 60, after attaching in a car body etc. -- conspicuous -- easy -- an exterior -- it is not desirable.

[0020] This L value is a scale showing a hue, and it measures in this invention by the color machine (SM color computer [by Suga Test Instruments Co., Ltd.] SM-5 mold). [0021] Although it really comes to fabricate a loft nonwoven fabric and polyolefine system resin foam, even if it uses cement for the lamination, it is not necessary to use the laminate of this invention. It is desirable to stick without using cement from recycle nature. As cement used in the case of the lamination of a laminate, heating fusion is carried out, and the adhesives of phenol resin, amino resin, an urethane system, a synthetic-rubber system, and an epoxy system, polyethylene, resin or fiber of polypropylene etc. that carries out reaction solidification may be mentioned, and these may be used independently, or may use together and use two or more kinds.

[0022] At a present stage, it is presumed as follows that a nonwoven fabric and foam

can be stuck on the other hand when not using cement, although it is not necessarily clear. In case a loft nonwoven fabric and polyolefine system bridge formation resin foam are really fabricated, on the other hand, both sheets are preferably heated in which. If these are really fabricated with lamination and a press, the surface part of a loft nonwoven fabric will sink into the surface part of polyolefine system resin foam, namely, an anchor effect-thing will be discovered, and it will be thought that it is stuck without both using cement.

[0023] For the thickness of the loft nonwoven fabric used by this invention, 5mm or more and a consistency are 50 kg/cm3. It is desirable that it is the following. Since a lappet etc. arises when thickness was thinner than 5mm, and the rigidity of mold goods runs short, for example, it uses for head-lining material etc., consistencies are 50 kg/cm3 preferably. In lamination with polyolefine system resin foam, a nonwoven fabric stops easily being able to sink into the surface part of polyolefine system resin foam as it is above, and the reinforcement of lamination becomes weak.

[0024] The skin peel strength of the polyolefine system resin foam used by this invention measured by the following approaches. Polyolefine system resin foam, the thickness of 1mm, and eyes 1.36~kg/m2 The test piece of a dimension with lamination, a width of face [of 25mm], and a die length of 100mm is created for the sheet of a vinyl chloride with adhesives. The reinforcement at the time of making a chlorination vinyl sheet this exfoliate at a room temperature using an autograph (product made from an incorporated company cage en tech "UCT500") is measured.

EXAMPLE

[Example] Next, although this invention is explained based on an example, this invention is not limited to this. That it is with the "section" below means the "weight section."

[0026] As manufacture polyethylene system resin of example 1(1) polyolefine system resin foam As the polyethylene 50 section whose MFRs are 5g / 10 minutes and whose melting point is 125 degrees C, and a polypropylene regin The ethylene propylene rubber 50 section in which ethylene carried out [MFR] random copolymerization 4.2% for 2.5g / 10 minutes, As a bridge formation assistant, as the divinylbenzene 3 section and a pyrolysis mold foaming agent The AZOJI carvone amide 12 section, The 2,6-di-t-butyl-p-cresol 0.3 section was supplied as an anti-oxidant, the carbon black 0.1 section was supplied to the biaxial extruder as a pigment, melting kneading was carried out at the temperature of 180 degrees C, and the fizz polyolefine system resin sheet with a thickness of 1.5mm was obtained. Carry out a 300kGy exposure, the electron ray was made to construct a bridge over the obtained fizz polyolefine system resin sheet as an ionizing radiation with the acceleration voltage of 800kV, and the fizz polyolefine system resin bridge formation sheet was obtained. The vertical mold foaming furnace was supplied, whenever [furnace temperature], the obtained fizz polyolefine system resin bridge formation sheet is continuously extended at 245 degrees C, and carried out heating foaming at them, and polyolefine system resin foam was obtained. Thus, one 25 times the expansion ratio [the thickness of 3.0mm, degree-of-cross-linking 35 %, and] of this and L value were [41 and the skin peel strength of the obtained foam | the things of 8.2 N/cm.

. (2) it the nonwoven fabric of 50mm in thickness which heated the manufacture profit **** polyolefine system resin foam of a layered product until skin temperature became 145 degrees C at the radiation heater, and heated the front face until it became 170 degrees C, and the polyester which is consistency 25 kg/m3, and really [lamination] fabricated, and the layered product was obtained.

[0027] In the manufacture of polyolefine system resin foam shown in the example of comparison 1 example, except not adding carbon black, the same actuation as an example 1 was performed, and one 25 times the expansion ratio [the thickness of 3.0mm, 35% of degrees of cross linking, and] of this and L value obtained the polyolefine system resin foam of 75, performed the still more nearly same actuation as an example, and obtained the layered product.

[0028] Instead of the nonwoven fabric of the polyester shown in the example of comparison 2 example, except having used the polyester nonwoven fabric of 50mm in thickness, and consistency 75 kg/m3, the same actuation as an example was performed and the layered product was obtained.

[0029] instead of [of the polyolefine system resin foam shown in the example of comparison 3 example] -- thick -- except that 3.0mm and expansion ratio used the polyolefine system resin foam of 5 times, 35% of degrees of cross linking, the L value 38, and skin peel strength 23.2 N/cm, the same actuation as an example was performed and the layered product was obtained.

[0030] About the layered product obtained in the above-mentioned example and the examples 1, 2, and 3 of a comparison, peel strength was measured, the adhesive property with a loft nonwoven fabric was evaluated, and the obtained result was shown in Table 1. The judgment of appearance nature trickled black ink into the polyolefine system resin foam of a laminate, made the case where it was conspicuous as x, and made O the case where that was not right.

[0031] The judgment of a desquamative state made O the case where x and adhesion were not checked in the case where the polyolefine system resin foam which carried out cohesive failure to the polyolefine system resin foam which foam was made to exfoliate and exfoliated at the room temperature on adhesion of the fiber of a loft nonwoven fabric or the front face of a loft nonwoven fabric is checked.

[0032]

[Table 1]

| | 外観性 | 剥離強度 | 剥離状態 |
|-------|-----|--------|------|
| | | (N/cm) | |
| 実施例 1 | 0 | 0.304 | 0 |
| 比較例 1 | × | 0.304 | 0 |
| 比較例 2 | 0 | 測定不能* | - |
| 比較例3 | 0 | 2.99 | × |

[0033] It is shown that front Naka and * are below measurement limitations.

[0034] The thing of an example was what it is lightweight, and fabrication is easy, is the beautiful laminate of an appearance, and is excellent also in recycle nature as $% \left(1\right) =\left(1\right) ^{2}$, compared with the example of a comparison so that clearly from Table 1.